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TITLE SINGLE COLUMN AND TWO-COLUMN H-D-T DISTILLATION EXPERIMENTS AT TSTA

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ABSTRACT

Cryogenic distillation experiments were performed at TSTA with H-D-T system by using a single column and a two-column cascade. In the single column experiment, fundamental engineering data such as the liquid holdup and the HETP were measured under a variety of operational conditions. The liquid holdup in the packed section was about 10 ~ 15% of its superficial volume. The HETP values were from 4 to 6 cm, and increased slightly with the vapor velocity. The reflux ratio had no effect on the HETP. For the two-column experiment, dynamic behavior of the cascade was observed.

1. INTRODUCTION

Separation of hydrogen isotopes by cryogenic distillation is a promising method for the mainstream fuel reprocessing and blanket tritium processing systems of fusion reactors. To establish and verify design and operation methods for cryogenic distillation columns, both computer-aided simulation and experimental studies are needed. A number of simulations[1][2] have been reported for the cryogenic distillation columns. Although those reports produced a great deal of valuable information, the experimental data for the cryogenic distillation columns previously published is still limited.

For the packed columns which are used for the cryogenic distillation, the HETP (Height Equivalent To a Theoretical Plate), the liquid holdup and the pressure drop are significant parameters in column performance. Factors which are expected to have influence on these parameters are as follows : vapor velocities within the column, reflux ratios, packing materials and physicochemical properties (density, viscocity and surfac tension) of fluids. Several workers have reported engineerin data for the cryogenic distillation column[3][4][5], however, n detailed discussion concerning the effect of the above-mentione factors was given in their reports, additionally, there are so differences among the results obtained and the cause of thi discrepancy has not been resolved yet. It is also essential tha separation characteristics of cascades composed of severa columns are experimentally examined. For these reasons, furth experimental studies are desirable.

The most significant purpose of the present study is to obtain fundamental engineering data for the cryogenic distillation column and to discuss the factors affecting the column performance. For this purpose, the HETP, the liquid holdup and the pressure drop were measured with the H-D-T system as functions of the vapor velocity, the reflux ratio and the physicochemical properties of fluids by using a column in the TSTA isotope separation system.[5] As the first stage of a research program dealing with cascade operations, dynamic behavior of two interlinked columns was also observed under varying operational conditions in the present study. The column cascade selected simulates a basic configuration of that proposed by the authors[6] for the blanket tritium processing systems. The information obtained from this study improves and confirms the ability to predict separation characteristics of actual-scale columns and cascades.

2. EXPERIMENTAL

2.1 Single column experiment

The single column experiment was performed with H-D and D-T systems under total reflux and total recycle modes. In the total recycle mode, the top and bottom streams are recycled through an equilibrator as illustrated in Fig. 1. The major role of the equilibrator is to promote the isotope exchange reaction (e.g. $H_2+D_2=2HD$). The lead column of the TSTA isotope separation system was used in the present experiment. The specifications of the lead column are presented in Table I. The refrigeration is supplied by helium gas, whose temperature can be lowered by a refrigerator to about 15~20 K, at the condenser and the packed section. The distillation column is enclosed within a thermal radiation shield cooled by liquid nitrogen in a vacuum jacket.

The experimental procedure is summarized as follows. The vacuum jacket and the distillation column were initially evacuated, and operation of the helium refrigerator was started. Hydrogen isotope gases were charged into the column from cylinders through an equilibrator for the H-D experiment. In the D-T experiment, uranium beds were used to supply the gases. The condensation of hydrogen isotopes occurred in the condenser and the packed section, and the liquid hydrogen thus produced fell to the reboiler. After the liquid level in the reboiler reached a specified value, the distillation was initiated. For the experiment with the D-T system, the hydrogen isotope mixture charged into the column was recycled through the equilibrator to make its composition the equilibrium state at room temperature before the distillation. Composition distributions within the column were measured by gas chromatographs after the column reached steady state.

2.2 Two-column experiment

The two-column experiment was performed with H-D-T using the total recycle mode. The configuration of the two-column cascade is shown in Fig. 2. The specifications of the second column are also presented in Table I. The general construction of the second column is the same as that of the lead column. The experiment was performed in a similar manner to that of the single column. After the gases were charged into the cascade, the columns were individually operated in the total reflux mode until steady state was achieved. The cascade was then put into total recycle operation. Dynamic variation of operating parameters such as the pressures and the liquid holdups were measured while varying the reflux ratio of each column.

3. Calculation of composition distribution within the column

The assumptions used in the present calculation are as follows :

(1) The column is composed of N theoretical stages.

(2) The column operates adiabatically, and the molar heats of vaporization of all components are the same.

(3) Hydrogen isotope mixture obeys Raoult's Law.

At the steady state, the component material balances are expressed by

$$0 = L_{N-1}x_{i,N-1} + F_N z_{i,N} - (V_N + W_N)y_{i,N} - L_N x_{i,N} , \qquad -(1)$$

$$0 = V_{j+1}y_{i,j+1} - (L_j + U_j)x_{i,j} + L_{j-1}x_{i,j-1} - (V_j + W_j)y_{i,j} + F_j z_{i,j} , \qquad -(2)$$

$$0 = V_2 y_{i,2} + F_1 z_{i,1} - (L_1 + U_1)x_{i,1} - V_1 y_{i,1} , \qquad -(3)$$

where F_j = flow rate of feed stream supplied to j-th stage (mol/h)

L_j = flow rate of liquid stream leaving j-th stage (mol/h)

N = number of total theoretical stages (-)

U_j = flow rate of liquid sidestream from j-th stage (mol/h)

V_j = flow rate of vapor stream leaving j-th stage (mol/h)

W_j = flow rate of vapor sidestream from j-th stage (mol/h)

$x_{i,j}$ = mole fraction of i-th component in the liquid stream leaving j-th stage (-)

$y_{i,j}$ = mole fraction of i-th component in vapor stream leaving the j-th stage (-)

$z_{i,j}$ = mole fraction of i-th component in feed stream supplied to j-th stage (-).

The vapor liquid equilibrium are expressed by

$$y_{i,j} = p_i(T_j)x_{i,j}/P_j \quad - (5)$$

where P_j = total pressure on j-th stage (Torr)

p_i = vapor pressure of i-th component (Torr)

T_j = temperature on j-th stage (K).

If the composition of either the top or bottom stream is given, Equation (1) through (3) can be solved by using the vapor-liquid relations expressed by Eq. (5).

The overall HETP value is determined from the packed height of the column and the number of total theoretical stages which gives the best fit to experimental observations. The simple model used is sufficient to determine the overall HETP value. Variation of the HETP with the column height can also be discussed by comparing experimental observations with calculational results for the composition distribution within the column.

4. RESULTS AND DISCUSSION

4.1 Pressure drop across column

A significant result observed is that the distillation was successfully performed without column flooding as long as the vapor velocity . less than 13 cm/sec. The relation between the pressure drops and the vapor velocity is shown in Fig. 4. For the total reflux experiment of the H-D system alone, the pressure drop is small and the loading might occur when the vapor velocity exceeds 12 cm/sec. For the other three experimental conditions, the measured values are proportional to the vapor velocity to the power 1.3~1.5. Although a firm conclusion on this difference has not been drawn yet, it may be ascribed to the hysterisis on the pressure drop. The total reflux experiment was performed by increasing the flow rates of each phase, and the column was then put into the total recycle operation. The increase of the vapor and liquid flow rates would affect wetting characteristics of the packing materials, and can possibly vary the fluidity within the column (i.e. variations in the amount of liquid held by the packing materials and in the dispersion mechanism of the liquid falling from the condenser.). For the D-T experiment, the packing materials may be adequately wetted owing to the recycle operation performed before the distillation.

4.2 Liquid holdup within packed section

The liquid holdup within the packed section can be evaluated by subtracting the measured value for the reboiler and calculated vapor holdups of the column from the total amount of gas charged. Figure 3 shows the liquid holdups thus obtained under a variety of vapor velocity conditions. The obtained values, which are about 10~15 mol, correspond to 10~15% of the superficial volume of the packed section. The values for the H-D system under the total reflux experiment are slightly smaller in comparison with those of the D-T sys . This difference appears to be due to the hysterisis discussed in the preceding section. For the other experimental conditions, no apparent difference is observed between the H-D system and the D-T system. As another significant result observed from Fig. 3, the liquid holdups for all the experimental conditions increase with the vapor velocity. For the above reasons, it can be concluded that the liquid holdup is a function of the vapor velocity.

4.3 Effects of vapor velocity and reflux ratio on HETP

Figure 5 shows the effect of the vapor velocity on the overall HETP values. For the total reflux mode, the values are about 4 5 cm, and those of the total recycle mode are from 5 to 6 cm. Sherman et al.(3) reported that the overall HETP values were approximately 5 ± 0.5 cm for the cryogenic distillation column whose inner diameter (0.95 cm) and packed height (45.7 cm) are considerably smaller. The overall HETP values measured in the present study for the actual size column are almost equal to that obtained by Sherman et al. For the total recycle mode, the separation performance is slightly deteriorated. Disturbance of the phase flow within the column caused by the recycle operation may result in the decrease of the vapor/liquid interfacial area. Comparing experimental results for the H-D system with those of the D-T system, we conclude that the physicochemical properties of fluids have no effect on the overall HETP value. For the total reflux experiment with the H-D system, the hysterisis was observed on the liquid holdup and the pressure drop. However, on the overall HETP, no hysterisis is observed.

Another significant result observed from Fig. 5 is that the overall HETP values for all conditions increase slightly with the vapor velocity. The overall HETP value can be expressed by[7]

$$\text{overall HETP} = z/(N-2), \quad (y_{i,1} - y_{i,N}) = \int_0^z (n_i/V_j)dz \, , \qquad -(6)$$

where n_i = mass transfer rate for component i ($molm^2/m^3hr$)

z = packed height of column (m).

The mass transfer rate n_i can be regarded as a function of the vapor velocity and the column height. If n_i increases proportionally to the vapor velocity, the overall HETP value should have no dependence on the vapor velocity. The present experimental results show that the mass transfer rate for the hydrogen isotope distillation system is proportional to less than the first power of the vapor velocity. Wilkes [4] obtained the smaller overall HETP values ($2\sim3$ cm) for the cryogenic distillation column in the vapor velocity range from 1 to 6 cm/sec. The difference in the vapor velocity may possibly be one of the reasons for the difference of the HETP values.

Figure 6 shows the effect of the reflux ratio on the overall HETP value when the vapor velocity and the flow rate of bottom stream are almost constant. The reflux ratio is varied from 8 to 35 ; the top stream ($\sim2\times10^{-3}$ mol/sec) is very small in comparison with the vapor flow rate (~0.028 mol/sec). In addition, the increment of the liquid flow rate is only 10 % of the initial value. The overall HETP values therefore would not be affected by the reflux ratio, as shown in Fig.6. The reflux ratio and the vapor velocity are suitable manipulated parameters of the distillation column to control purities of top and bottom products at desirable values.[8] The model used in the present study has the disadvantage that it is difficult to consider the variation of the number of total theoretical stages with operating conditions when dynamic behavior of the columns under control mode is simulated.[2] However, the constancy of the overall HETP against the reflux ratio and its slight dependency on the vapor velocity indicate that the simulation results by the stage model can be extended to actual column behavior even for the control operation.

As previously mentioned, the overall HETP value was determined without regard to the composition distribution within the column. Figure 7 shows the composition distribution at the steady state for a representative run. The calculated lines are also drawn in the figure under the assumption that HETP is constant with column height. Table II shows the experimental and calculational conditions of the run. The composition distribution calculated is in rough agreement with experimental observation. The HETP depends little on the column height. The above conclusion is important for column design : positions of feed and side-cut streams of actual columns can be well estimated by the stage model.

4.4 Dynamic behavior of column cascade

This section will be added to the present manuscript after experimental results of the two-column are obtained.

Figure : 1 or 2

Text : about 1p

5. CONCLUSION

(1) In the range of the vapor velocity to 13 cm/sec, there was no evidence that the flooding occured. The hysterisis observed on the pressure drop across the column appears to be mainly related the variation of the fluidity within the column.

(2) The liquid holdup in the packed section was approximately 10 15 % of its superficial volume, and increased with vapor velocity.

(3) The measured overall HETP values were about 4 5 cm for the total reflux mode, and were from 5 to 6 cm for the total recycle mode. The overall HETP increased slightly with vapor velocity, and showed no dependence on the reflux ratio or the physicochemical properties of the fluid. The HETP was relatively constant within the column.

(4) The stage model is well suited for the simulation of actual column behavior and performance.

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Table I Specifications of distillation columns

	lead column	second column
Inner diameter (cm)	2.84	2.50
Packed height (cm)	412	320
Volume of condenser (cm^3)	2000	2000
Volume of packed section (cm^3)	2600	1600
Packing material	Heli-Pak (SUS-316) 4.4x4.4x2.3 mm	

Table II Experimental and calculational conditions

Operation mode		total recycle
Operating pressure (Torr)		770
Flow rate (mol/sec)	top stream	1.49×10^{-3}
	bottom stream	1.68×10^{-3}
Vapor velocity (cm/sec)		5.65
Liquid holdup of reboiler (mol)		4.44
Number of total theoretical stages		86
Feed stage number		60

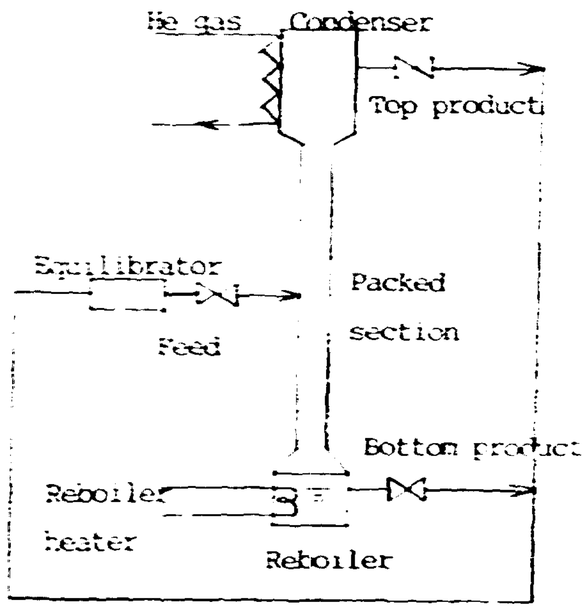

Fig. 1 Conceptual flow diagram
for single column experiment

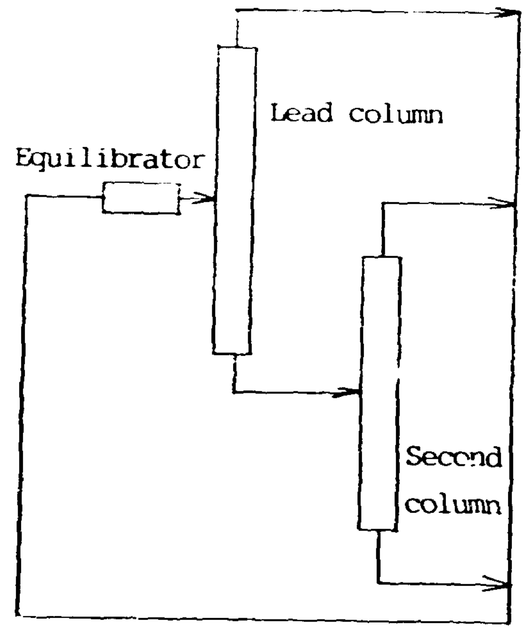

Fig.2 Configuration of cascade

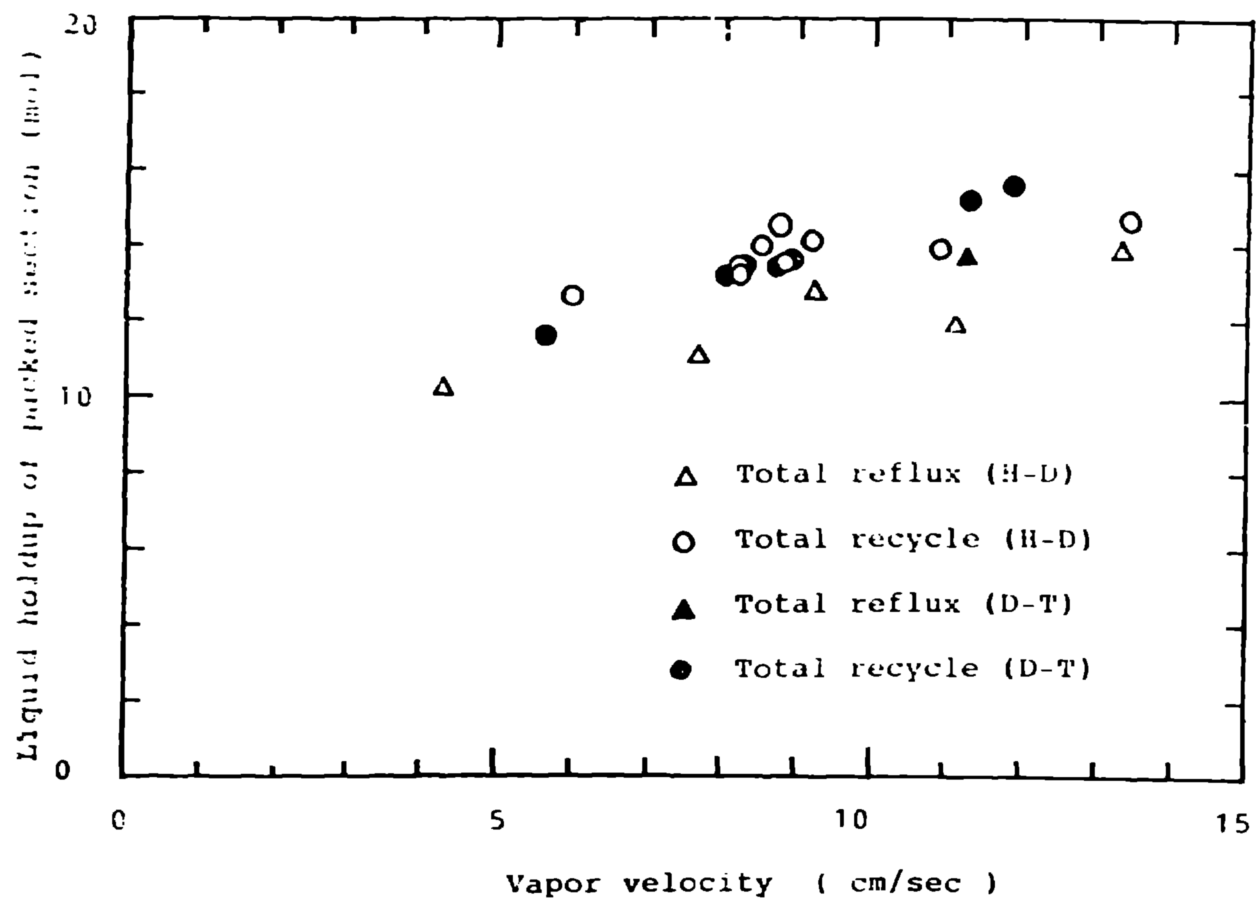

Fig. 3 Variation of liquid holdup in packed section with vapor velocity .

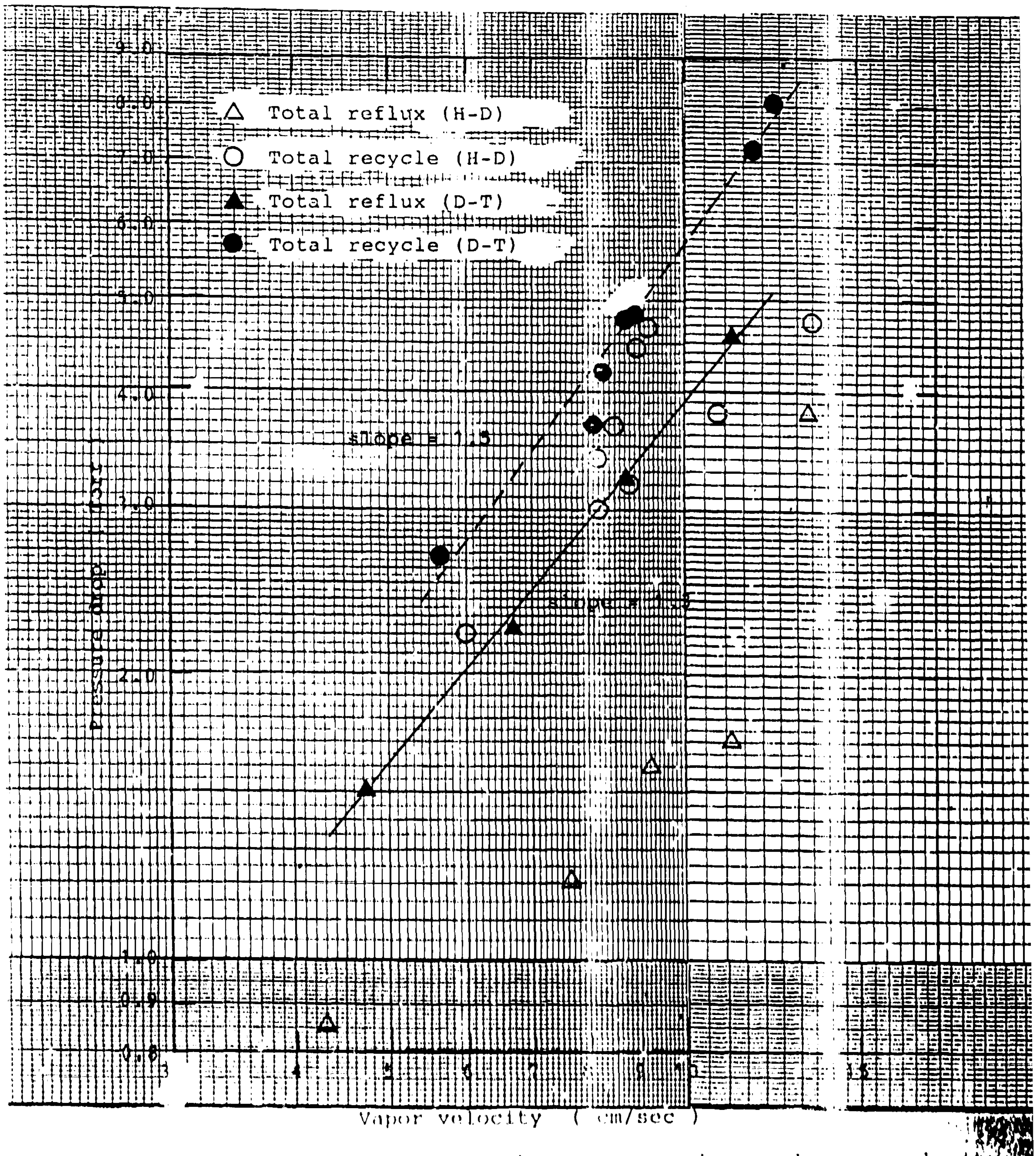

Fig.4 Relation between pressure drop across column and vapor velocity

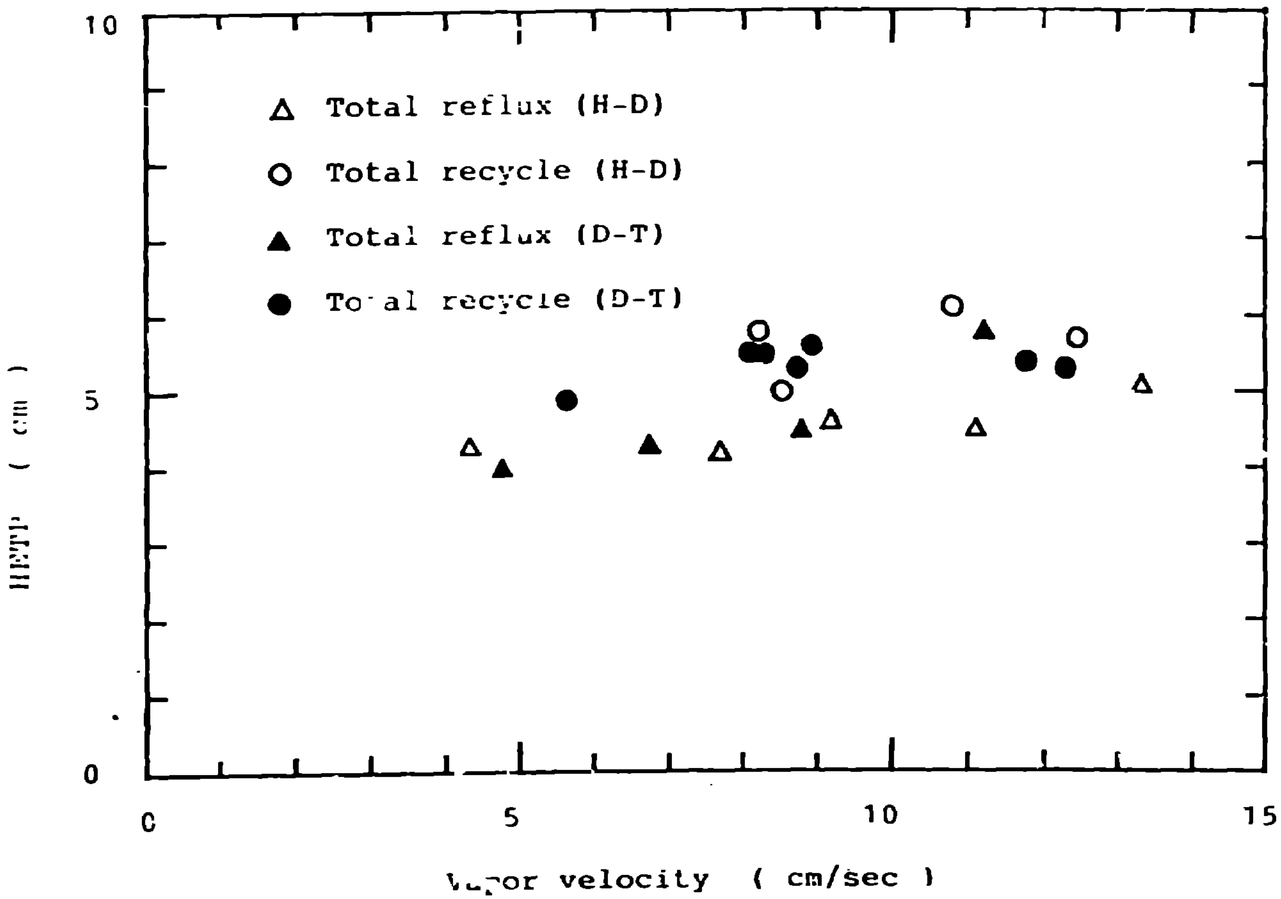

Fig. 5 Effect of vapor velocity on overall HETP value

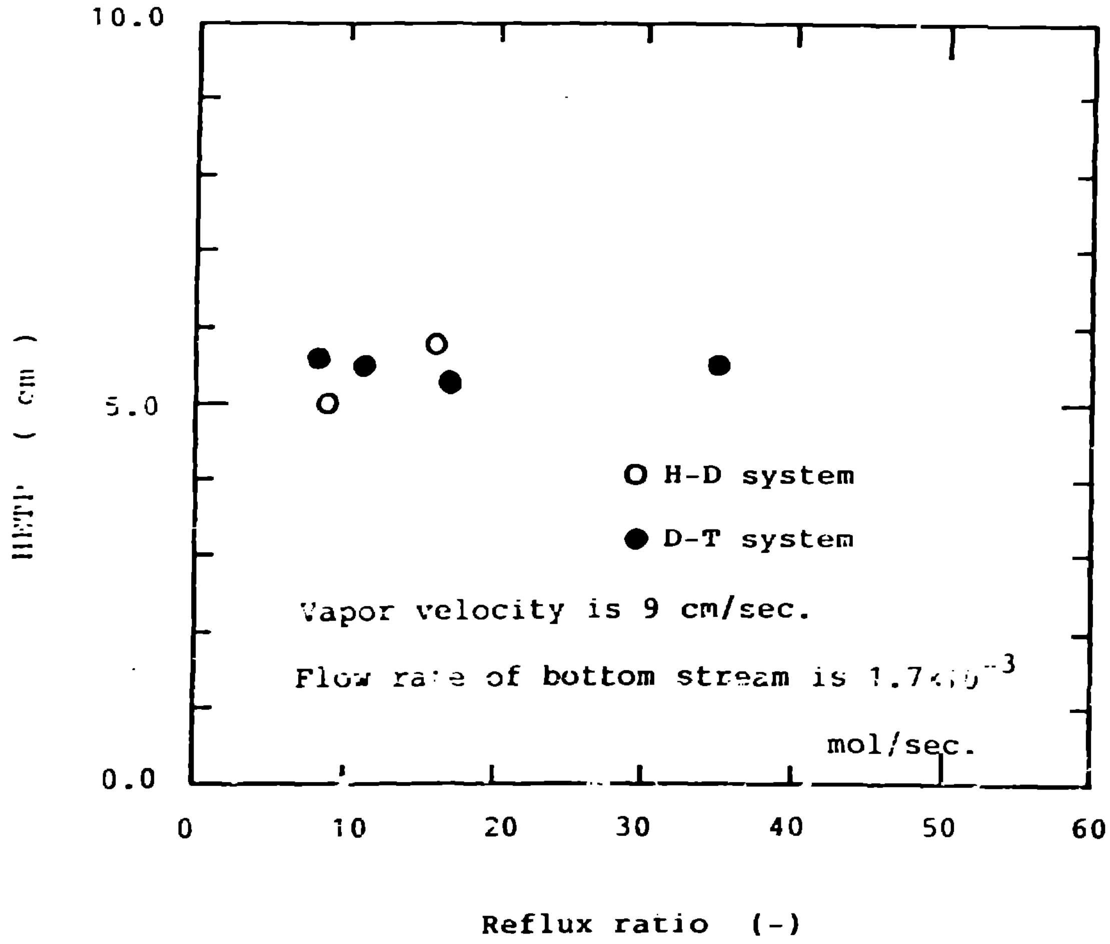

Fig. 6 Effect of reflux ratio on overall HETP value

MODEL DATE

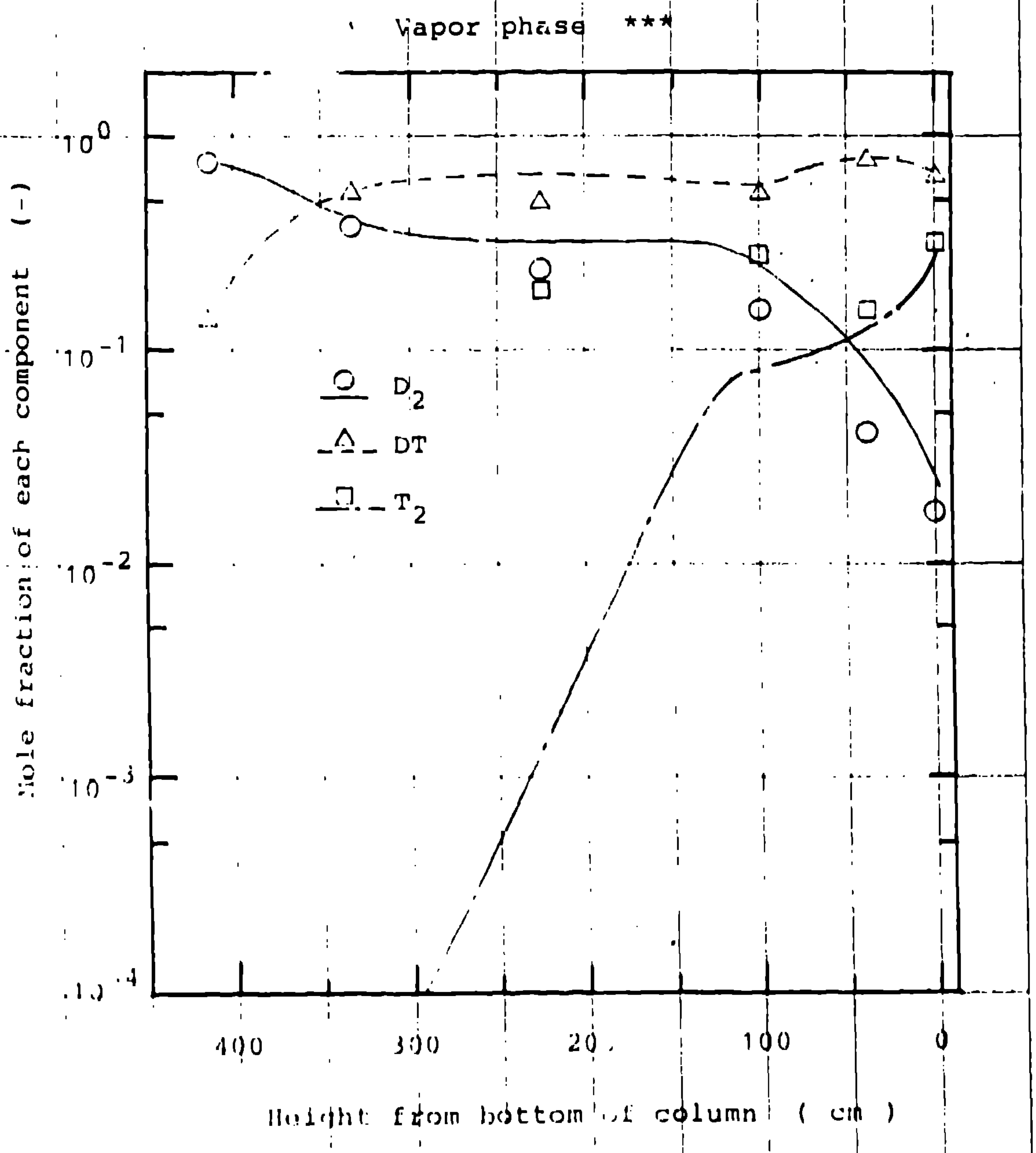

Fig. 7 Comparison between experimental observation and calculational result for composition distribution